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Effect of Annealing Temperature on Low-temperature Toughness of *β*-Nucleated Polypropylene Random Copolymer/Ethylene-Propylene-Diene Terpolymer Blends*

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Abstract In this study, the effect of annealing temperature on the impact toughness of *β*-nucleated polypropylene random copolymer (PPR) and ethylene-propylene-diene terpolymer (EPDM) blends was investigated by differential scanning calorimetry (DSC), wide-angle X-ray diffraction (WAXD), dynamic mechanical analysis (DMA) and scanning electron microscopy (SEM). Interestingly, the impact strength of *β*-PPR/20EPDM blend annealed at 120 °C is 1.8 times as high as that of unannealed samples. In addition, the crystalline structure, the relaxation of chain segments and fracture morphology of *β*-PPR/EPDM blends were also investigated to explore the toughening mechanism related to annealing. The results show that annealing at moderate temperatures results in the improvement of integrity of the crystal structure and the relative content of *β*-phase. The work provides a possible method to toughen the semicrystalline polymer at low temperatures by annealing.

Keywords: Polypropylene; EPDM; *β*-Nucleating agent; Annealing; Toughening.

INTRODUCTION

Polypropylene (PP) is one of the most widely used thermoplastics with numerous advantages, such as low density, easy processing, low cost and good mechanical properties; but its application in some fields is limited by its brittleness especially at low temperatures, as well as its sensitivity to notch. In order to improve the impact toughness of PP and extend its application range, numerous methods have been proposed in the past several decades, mainly including copolymerization and blending[1–5]. Copolymerization of propylene with ethylene or other olefins is an effective method to produce high-performance polypropylene resins. In such copolymers, the propylene-ethylene random segments with high ethylene contents coalesce together to form a rubbery phase; while the homopolymer sequences are crystallizable to form a compound crystalline-amorphous biphase. Therefore polypropylene random copolymer (PPR) is indeed a multiphase polyolefin system with an excellent rigidity-toughness balance: the crystalline phase of homopolymer sequences guarantees the moderate strength and modulus, and the well-dispersed rubbery domains offer superior toughness[1, 3]. PPR materials have been widely used in both domestic and industrial applications in the past two decades, based on the excellent mechanical properties and relatively low cost[6]. Even so, the toughness of PPR at low temperatures (below T_g) is still too low to limit its further applications. To improve the impact toughness of PPR, the blends of PPR and elastomeric modifiers such as ethylene-propylene-diene terpolymer (EPDM), styrene-butadiene-styrene (SBS)

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and ethylene-propylene rubber (EPR) have been one of the most commercially successful impact modified polymer systems. Among these rubbers, EPDM is considered as one of the most effective impact modifiers for PPR[7–9].

It is well known that polypropylene is a polymorphic material with at least four crystalline forms, namely, monoclinic α-form, trigonal β-form, orthorhombic γ-form, and mesomorphic smectic form[10, 11]. Under standard synthesis and processing conditions, polypropylene generally crystallizes into the α-phase, while β-phase can be prepared by the introduction of specific nucleating agent, *i.e.* β nucleating agent (β-NA), which is the most effective and accessible method to obtain PP with high content of β crystals[3, 8]. The differences of supermolecular structures and the different crystalline states of PP exhibit different mechanical features. For example, the α-phase with a typical cross-hatched lamella structure and compact stacking of molecular chains endows excellent modulus and tensile strength to PP, but shows inferior fracture toughness because the presence of interlocking effect of the radial lamellae by tangential crystallites makes the plastic deformation very difficult[12, 13]. On the contrary, the loose structure of β-crystals makes the initiation and propagation of plastic deformation very easy and then enhances the energy dissipation[3, 14, 15]. However, the presence of β-phase has little effect on the low-temperature (below T_g) toughness of polypropylene[16]. Currently, blends of rubber particles and β-modification have been considered to largely improve fracture resistance of polypropylene and promote the shift of brittle-ductile transition to lower rubber particle volume fractions[14, 17].

Recently, the effect of annealing on the structure and properties of polymers has been studied extensively. Because it is difficult for polymer chains to achieve the equilibrium state and generally imperfect packing structures are formed when polymer melt is rapidly cooled down from the melt temperature to room temperature in polymer processing. Nevertheless, annealing at moderate temperature between glass transition temperature (T_g) and melting temperature (T_m) is thought to be an effective method to promote chain relaxation toward a thermodynamically more stable state and induce significant changes in microstructure and physical properties of polymers[18]. Some works have proved that fracture resistance of PP annealed at proper conditions can be notably improved due to microstructure changes by secondary crystallization, such as the perfection of defective crystals, thickening of crystal lamella, partial melting-recrystallization, phase transition of polymorphs and molecular rearrangement[19, 20]. Although a great amount of work has been carried out to study the effect of annealing on the structures and properties of PP or PP blends, the effects of annealing on impact toughness of β-PPR/EPDM at low temperatures have not been specially investigated up to now.

In this work, we attempted to further improve the impact toughness of β-PPR/EPDM blends at relatively low temperature. So we focus on the effects of microstructure evolution for β-PPR/EPDM on mechanical properties, melting behavior and morphologies during annealing at different temperatures, and deeply understand the structure-property relationship of PP based materials.

EXPERIMENTAL

Materials

Commercial grade polypropylene random copolymer (PPR, trademark R200P) with $M_w = 7.22 \times 10^5$ g/mol, $M_w/M_n = 4.5$, a melt flow rate (MFR) of 0.27 g/10 min (230 °C, 2.16 kg load) and density of 0.89 g/cm^3 was supplied by Hyosung (Korea). The mass percentage of ethylene component was as low as 3.8 wt%. EPDM was a commercial grade 3722P supplied by Dow Chemicals (Washington, USA), with $M_w = 10^5$ g/mol and density of 0.88 g/cm^3. The mass percentage of ethylene and ethylidene norbornene (ENB) component was 71 wt% and 0.5 wt% respectively. The rare earth β-nucleating agent, marked as WBG, was kindly supplied by Winner Functional Materials Co. (Foshan, Guangdong, China). A small amount of antioxidant (Irganox 1010) was added into blends to prevent the thermal decomposition during melt processing.

Sample Preparation

PPR containing 0.12 wt% β-nucleating agent named as β-PPR and β-PPR containing 20 wt% EPDM named as β-PPR/20EPDM were prepared respectively by melt blending in an SHJ-38 co-rotating twin-screw extruder with the setting temperatures of 180–205 °C from hopper to die and the screw speed of 200 r/min. The extrudate was then quenched into water and pelletized. After drying to remove the moisture at 80 °C for 4 h, the pellets were injection-molded into standard samples for testing on a HTF86X1 precise injection-molding machine, with a temperature profile of 190–210 °C from the feeding zone to the nozzle and injection pressure of 40 MPa.

The annealing of the samples at selected annealing temperatures (T_a) for 1 h was performed in a temperature-controlled oven. After annealing, the samples were cooled in the ambient air and kept at 23 °C for at least 24 h before testing.

Mechanical Test

The standard tensile test was performed on a dumbbell-shaped sample using an XWW-20 testing machine (Chengde Jinjian Testing Instrument Co., Ltd., Hebei, China) at ambient temperature (23 °C) according to ASTM D638-03. The crosshead speed of 100 mm/min was used. The flexural test was conducted on a WSM-20KN universal materials testing machine (Changchun Intelligent Instrument Equipment Co., Ltd., Jilin, China) with a loading speed of 2 mm/min according to ASTM D790. The average values were obtained over five samples by the tensile and flexural testing respectively. The notched Izod impact strength of the samples was measured with a JJ-5.5J Izod machine (Changchun Intelligent Instrument Equipment Co., Ltd., Jilin, China) according to ASTM D256-04. For low temperature impact tests, the samples were kept for at least 4 h at −30, −25, −20, −15, −10 and 0 °C, respectively and then immediately subjected to impact. At least twenty samples were tested and the average values were reported.

Differential Scanning Calorimetry (DSC)

The thermal behavior of samples were measured on a Perkin-Elmer Diamond DSC calorimeter (USA) in a nitrogen atmosphere. For each measurement, a 5–10 mg sample sealed in aluminum pan was heated from 50 to 200 °C at a rate of 10 K/min, and the DSC traces of first heating scan were reported.

Wide-angle X-ray Diffraction (WAXD)

WAXD measurements were carried out with a D/max-2500/PC X-ray diffractometer (Japan) with nickel-filtered CuKα radiation (λ = 0.154 nm). The patterns were recorded in the diffraction angle (2θ) range of 5–50° at 40 kV and 40 mA with a scanning velocity of 6 (°)/min. The relative content of β-PPR (K_β) in the crystalline phase was evaluated by the method proposed by Turner-Jones *et al.*[21]:

$$K_\beta = \mathrm{H}_{300}^{\beta} / (\mathrm{H}_{110}^{\alpha} + \mathrm{H}_{040}^{\alpha} + \mathrm{H}_{130}^{\alpha} + \mathrm{H}_{300}^{\beta}) \tag{1}$$

where $\mathrm{H}_{110}^{\alpha}$, $\mathrm{H}_{040}^{\alpha}$, and $\mathrm{H}_{130}^{\alpha}$ are the integral intensities of (110), (040) and (130) diffraction peaks of α-PP, respectively, and H_{300}^{β} is the integral intensity of (300) diffraction peak of β-PPR.

Dynamic Mechanical Analysis (DMA)

DMA testing was performed using a Tritec 2000 dynamic mechanical analyzer (Triton, Britain). Samples with a rectangular shaped part in the size of 15 mm × 10 mm × 4 mm were used to perform the test under dual cantilever bending mode from −100 to 100 °C in nitrogen atmosphere at a vibration frequency of 1 Hz and a heating rate of 3 K/min.

Scanning Electron Microscopy (SEM)

The SEM experiments were performed using a 1350VP SEM instrument (LED, Germany) with an acceleration voltage of 20 kV. For the observation of impact fracture surface morphology, EPDM were selectively etched with xylene (2 h) and then washed by pure water. Before observation, all the impact fractured surfaces were coated with gold.

RESULTS AND DISCUSSION

The Effect of Annealing on Fracture Toughness of β-PPR/EPDM

On the basis of the research goal of this study, the effect of annealing temperature (T_a) on the notch Izod impact strengths of *β*-PPR and *β*-PPR/EPDM at −20 °C was investigated. As shown in Fig. 1, the impact strengths of *β*-PPR annealed at different temperatures do not change distinctly and are still poor. However, the sample of *β*-PPR/20EPDM annealed at 120 °C shows obviously high impact strength (41.2 kJ/m^2), which is about 1.8 times as high as that of unannealed *β*-PPR/20EPDM (23.6 kJ/m^2) and about 11.7 times as high as that of *β*-PPR annealed at 120 °C (3.5 kJ/m^2). Moreover, the impact strength of *β*-PPR/20EPDM annealed at 130 °C (30.9 kJ/m^2) is lower than that of the one annealed at 120 °C. For *β*-PPR/20EPDM blend, the toughening effect of annealing treatment at 120 °C is obviously higher than that at other temperatures (≤ 130 °C), implying that the improvement in toughness is strongly dependent on annealing temperature owing to the microstructure changes of *β*-PPR/20EPDM blend during annealing process. In addition, the brittle-ductile transition temperature of *β*-PPR/20EPDM annealed at 120 °C (about −20 °C) is lower than that of the unannealed one (about −15 °C), as shown in Fig. 2. In a word, EPDM and annealing treatment exhibit synergistic effect in toughing *β*-PPR. Furthermore, the synergistic effect is greatly dependent upon the annealing temperature.

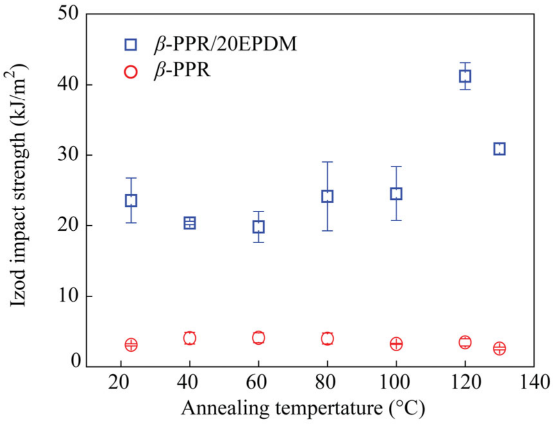

Fig. 1 The notch Izod impact strengths of *β*-PPR and *β*-PPR/20EPDM annealed at different temperatures, tested at −20 °C

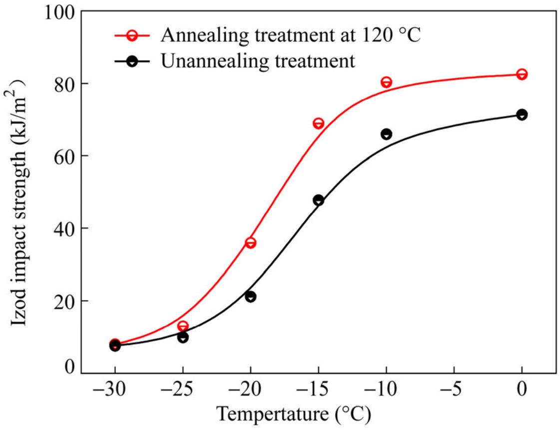

Fig. 2 The influence of annealing treatment on brittle-ductile transition of *β*-PPR/20EPDM

The tensile and flexural properties of *β*-PPR and *β*-PPR/20EPDM annealed at different temperatures were also studied, and the results are shown in Table 1. It is found that the yield strength and flexural strength of *β*-PPR and *β*-PPR/20EPDM increase when the annealing temperature increases. Furthermore, at low annealing temperatures (< 80 °C), the tensile elongation of samples is very high, while the tensile elongation decreases greatly at high annealing temperatures (≥ 80 °C). It is interesting to notice that the variation trends of impact strength and tensile elongation are different when the annealing temperature increases. The similar results have been reported in some works[1, 17]. Generally, it is recognized that impact strength and tensile elongation can both represent the toughness of materials. However, impact strength and tensile elongation don't always vary in the same trend due to their different deformation conditions. Impact strength represents the impact resistance of materials under high deformation rate condition, during which the materials are unable to response to the load promptly, while tensile elongation represents that under low deformation rate conditions, during which the materials are able to response promptly. Furthermore, during impact fracture process, the cavitations can trigger the plastic deformation and then promote energy dissipation to improve the impact fracture toughness of polymers. On the contrast, during tensile test process, the much lower deformation rate compared to that in impact fracture process makes crystalline elements deform plastically before reaching the cavitation threshold, which strongly prohibits the cavitation process. Moreover, during tensile fracture process, the microvoids can develop into microcracks, which has largely deteriorated the elongation[1, 22]. Thus, annealing treatment makes the impact strength of samples increase greatly but the elongation decrease largely.

Table 1. Tensile and flexural properties of *β*-PPR and *β*-PPR/20EPDM annealed at different temperatures

Annealing temperature (°C)	*β*-PPR			*β*-PPR/20EPDM		
	Yield strength (MPa)	EB[a] (%)	Flexural strength (MPa)	Yield strength (MPa)	EB[a] (%)	Flexural strength (MPa)
23	26.1	> 533.7	17.1	17.3	> 504.7	10.1
40	26.7	> 539.7	17.5	18.3	> 504.4	10.1
60	27.1	> 523.6	17.4	18.7	> 506.6	9.9
80	28.6	103.9	17.2	19.6	392.6	9.8
100	28.7	81.5	18.0	20.3	343.9	10.5
120	28.7	47.5	18.7	20.7	318.8	10.9
130	28.2	165.4	19.9	21.1	304.0	11.8

[a] Tensile elongation; > no tensile fracture in the given process

The Effect of Annealing Temperature on the Crystalline Structure for β-PPR/EPDM

Figure 3 shows the DSC melting curves of *β*-PPR/20EPDM annealed at different temperatures. After annealing at low temperatures (≤ 100 °C), *β*-PPR/20EPDM exhibits a shoulder peak in the thermogram before the main fusion peak of *α*-PPR and *β*-PPR, and the onset temperature of the shoulder peak increases gradually with the increase of the annealing temperature, indicating that annealing induces a microstructure change, which is dominated by the applied annealing temperature, as plotted in the inset of Fig. 3. It is known that the endothermic shoulder peak is attributed to the melting of thin and imperfect lamella formed during the secondary long-time annealing process, and similar results have been reported in some literatures[20, 23]. Moreover, annealing at the temperature higher than the upper limit of nucleation of *β*-phase for *β*-PP results in an increase of the average lamellar thickness and narrowing of the distribution[24]. As shown in Fig. 4, the annealing temperature of 120 °C is higher than the upper limit of crystallization temperature of *β*-phase for *β*-PPR/20EPDM (116.4 °C). Thus, when the annealing temperature is increased from 23 °C to 120 °C, the onset temperature of the melting peak of *β*-PPR is increased by about 20 K (from 95.1 °C to 116.4 °C) and the half width is decreased by 4.1 K (from 12.3 °C to 8.2 °C), indicating that the lamellae of crystals are thickened and the distribution of the lamella thickness is narrowed owing to the crystal perfection process during annealing.

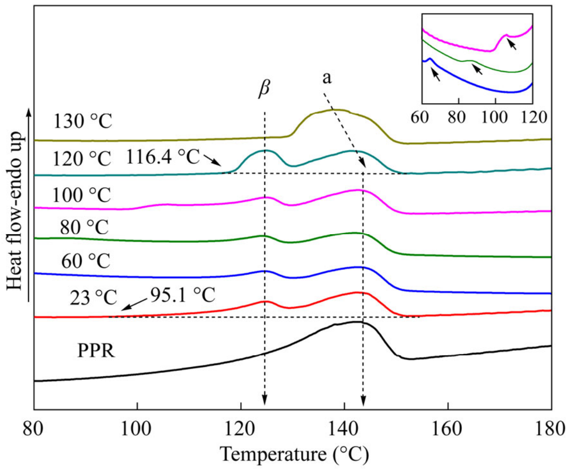

Fig. 3 DSC melting curves of *β*-PPR/20EPDM annealed at different temperatures

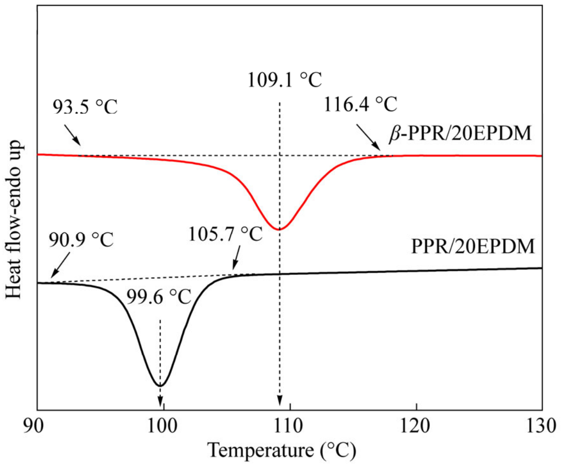

Fig. 4 DSC cooling curves of PPR/20EPDM and *β*-PPR/20EPDM

Interestingly, the fusion peak of *β*-PPR disappears and only the fusion peak of *α*-PPR is observed at annealing temperature of 130 °C, indicating that at the much higher temperature compared to the melting temperature of *β*-PPR (about 125 °C), the annealing makes the unstable *β*-PPR melt and recrystallize as *α*-PPR with more stable crystalline structure, namely, a transformation of *β*-PPR to *α*-PPR occurs during the annealing process. It is believed to be favorable to the improvement of yield strength and flexural strength as shown earlier.

The crystalline structure of *β*-PPR/20EPDM annealed at different temperatures was also investigated by WAXD and the relative fraction of *β*-PPR was calculated by the Turner-Jones method[21] as shown in Fig. 5. For unannealed *β*-PPR/20EPDM blend, the relative fraction (K_β) of *β*-PPR is 11.9%, whereas the K_β of *β*-PPR is increased up to about 16% at annealing temperatures of 100 °C and 120 °C, which may be ascribed to the improvement of the crystalline structure of *β*-PPR during annealing process. Moreover, the grown-up lamellae of *β*-PPR might also expose the self-aggregated *β*-nucleating agents in PPR matrix during annealing process, leading to more *β*-PPR formed.

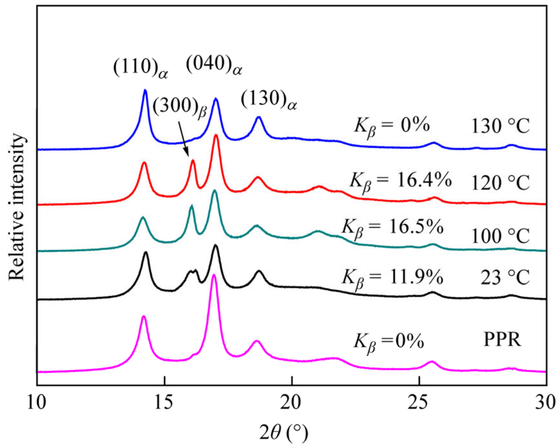

Fig. 5 WAXD patterns of *β*-PPR/20EPDM annealed at different temperatures

For semi-crystalline polymers, the size and the integrity of spherulites, as well as the boundary strength between spherulites, can influence the fracture resistance, namely, the fineness of the crystal structure is beneficial for the improvement of fracture resistance[25]. Moreover, the presence of *β*-phase can improve the molecular entanglement between the spherulites, thus increasing the boundary strength between the spherulites, then the fracture resistance is improved[26]. Thus, the reason for the increasing of the fracture resistance at annealing temperature of 120 °C is attributed to the improvement of the integrity of the crystal structure and the increase of the content of *β*-PPR.

The Effect of Annealing on the Amorphous Phase of β-PPR/EPDM

It is well known that the fracture resistance is the substantial response of molecular chain mobility. Jafari *et al.*[27] found a direct correlation between impact properties and dynamic mechanical loss tangent, thus the energy dissipation due to viscoelastic relaxation is suggested as a mechanism for toughening PP. As illuminated by this, the structure relaxation of unannealed and annealed samples was detected by DMA, and the results are shown in Fig. 6.

It can be seen from Fig. 6 that mechanical loss factor (tan*δ*) curves of *β*-PPR/20EPDM exhibit three maxima. The maximum at about −40 °C is related to *α*-relaxation, accounting for the glass transition of EPDM; the maximum at about 5 °C is related to *β*-relaxation, accounting for the glass transition of unrestricted amorphous PPR (T_g); and the maximum at about 70 °C is related to the α_c-relaxation, accounting for the relaxation of restricted PPR amorphous chains in the crystalline phase (defects)[28, 29]. The T_g value of the sample annealed at 120 °C moves to lower temperatures compared with that of the unannealed one, indicating that annealing at 120 °C promotes the mobility of PPR amorphous chain segments. Some works[20, 30] pointed out that this may be ascribed to a decrease in the concentration of the chain segments in the amorphous phase due to the second crystallization process relating to the change of some chain segments from amorphous state to crystalline state during annealing process. In addition, the low T_g value of samples annealed at 120 °C may be also attributed to the improvement of the relative content of β-PPR with loose crystalline structure. However, the T_g value of the sample annealed at 130 °C does not change compared with the unannealed sample, which is attributed to the transition of β-phase to α-phase. The presence of α-phase with dense cross-hatching structure makes the molecular chain mobility difficult, thus leading to the deterioration of fracture resistance. Furthermore, the intensity of β-relaxation of the sample annealed at 120 °C exhibits higher value than that of the unannealed sample. As is well known, the intensity of β-relaxation is representative of the maximum energy dissipated because of the viscoelastic relaxation of PPR components. This means that after being annealed at 120 °C, the fracture resistance and damping capacity are improved simultaneously.

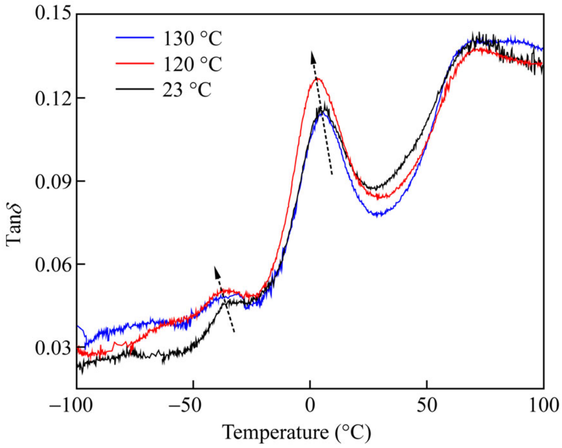

Fig. 6 Mechanical loss factor (tanδ) as a function of temperature for β-PP/20EPDM

The Effect of Annealing on the Fracture Morphology of β-PPR/EPDM

To clearly understand the variation of fracture resistance during the annealing process, the typical impact-fractured surfaces of β-PPR/20EPDM blend before and after being annealed at 120 °C and 130 °C were characterized, and the results are shown in Fig. 7. As shown in Figs. 7(a_1), 7(b_1) and 7(c_1) at low magnification, the impact-fractured surface of the unannealed sample appears to be relatively smooth, whereas relatively coarse impact-fractured surface with visible plastic deformation is observed in annealed samples. This suggests that annealing changes the fracture mode of β-PPR/20EPDM from brittle to ductile fracture, causing more energy dissipation by plastic deformation during impact fracture process. In Figs. 7(a_2), 7(b_2) and 7(c_2) at high magnification, the dark holes represent the EPDM particles, which were removed from the matrix by xylene. Clearly, the coarser impact-fractured surface with more visible shear deformation of the sample annealed at 120 °C can be observed compared with the sample annealed at 130 °C, which is attributed to their different crystalline structure. Wu[31] differentiated the fracture mechanism during impact tests depending on the nature of matrix, and when the matrix is brittle, the main fracture mechanism occurs by matrix crazing, whereas in the case of ductile matrix, the main fracture mechanism is by matrix shear yielding. Due to more β-PPR formed during annealing at 120 °C, the toughness of the matrix is improved and meanwhile the toughening effect of EPDM is enhanced, thus the large shear deformation takes place during fracture process, leading to the improvement of fracture resistance.

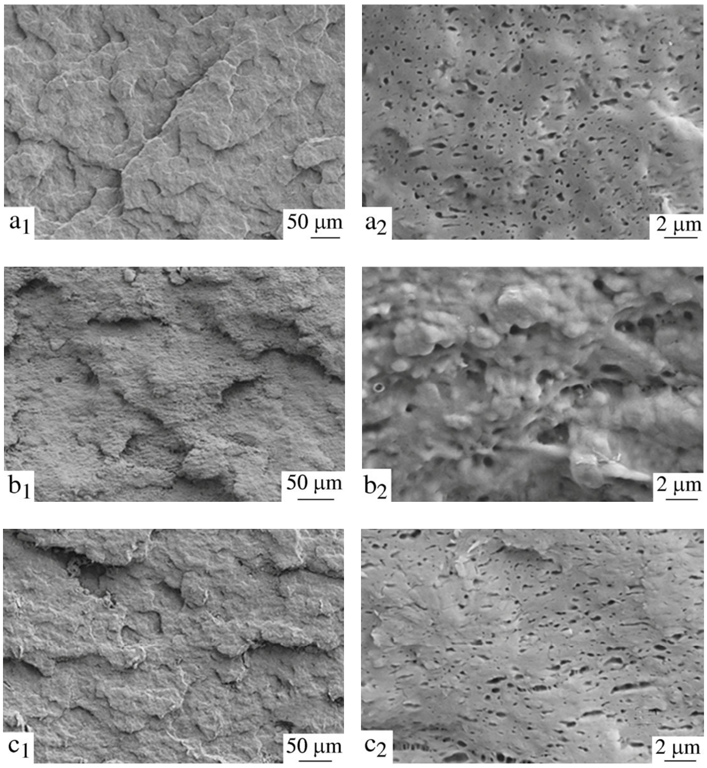

Fig. 7 SEM micrographs of the impact-fractured surfaces of *β*-PPR/20EPDM blends: (a_1, a_2) without annealing, (b_1, b_2) with annealing at 120 °C and (c_1, c_2) with annealing at 130 °C (a_1, b_1, c_1) are at low magnification, (a_2, b_2, c_2) are at high magnification. (fractured surface was treated with xylene at 23 °C, 2 h)

CONCLUSIONS

The improvement of low-temperature toughness of *β*-PPR/EPDM blends is strongly dependent on the annealing temperature. The impact strength of *β*-PPR/20EPDM blend annealed at 120 °C is improved greatly and about 1.8 times as high as that of unannealed *β*-PPR/20EPDM, which is attributed to the microstructure changes of *β*-PPR/20EPDM blend during annealing process. On the one hand, annealing at temperatures higher than the upper limit of nucleation of *β*-phase results in an increase of the average lamellar thickness and narrowing of the distribution, thus the improvement of the integrity of the crystal structure is beneficial for the improvement of fracture resistance. On the other hand, annealing at moderate temperatures improves the relative content of *β*-PPR, leading to the improvement of toughness of PPR matrix. In brief, our work proves a considerable way to increase the low temperature toughness of *β*-nucleated semicrystalline polymer by improving the integrity of the crystal structure and the relative content of *β*-phase.

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